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Robert A. Taft Sanitary Engineering Center
Cincinnati, Ohio

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RADIOACTIVE PARTICLES IN THE CINCINNATI ATMOSPHERE

February 16 - May 12, 1955

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INTRODUCTION

The radioactivity of air particles in samples collected from 20 to 30 municipalities in the National Air Sampling Network, established in 1953, indicated an increase of beta radioactivity of 1000 fold or more immediately following known atomic weapon tests. It was suspected that substantial amounts of the total radioactivity might be associated with discrete particles which might find their way into the respiratory tract of man. The purpose of the study was to determine the radioactivity distribution of collected particles. Samples collected at the Robert A. Taft Sanitary Engineering Center, Cincinnati, Ohio, during the spring of 1955 provided the test material for this study, the results of which are reported herein.

SAMPLING APPARATUS

The particles in air samples were separated into primary and secondary fractions by an Aerotec tube in series with a filter as shown in Figure 1. An air sampling rate of approximately 40 cubic feet per minute was selected, since at this flow the Aerotec tube (design 2, Thermix Corporation) has an efficiency vs. particle size curve comparable to that of the upper human respiratory system (3, 10).

A nosepiece on the intake of the Aerotec tube provided a face velocity of 25 liters per minute per square centimeter which approximates the nasal air velocity of a man at work. Particles passing the Aerotec were caught on the glass fiber filter and constituted the secondary fraction or that which might have penetrated to the alveoli in actual breathing. These particles were deposited on an effective filter area of 63 square inches.

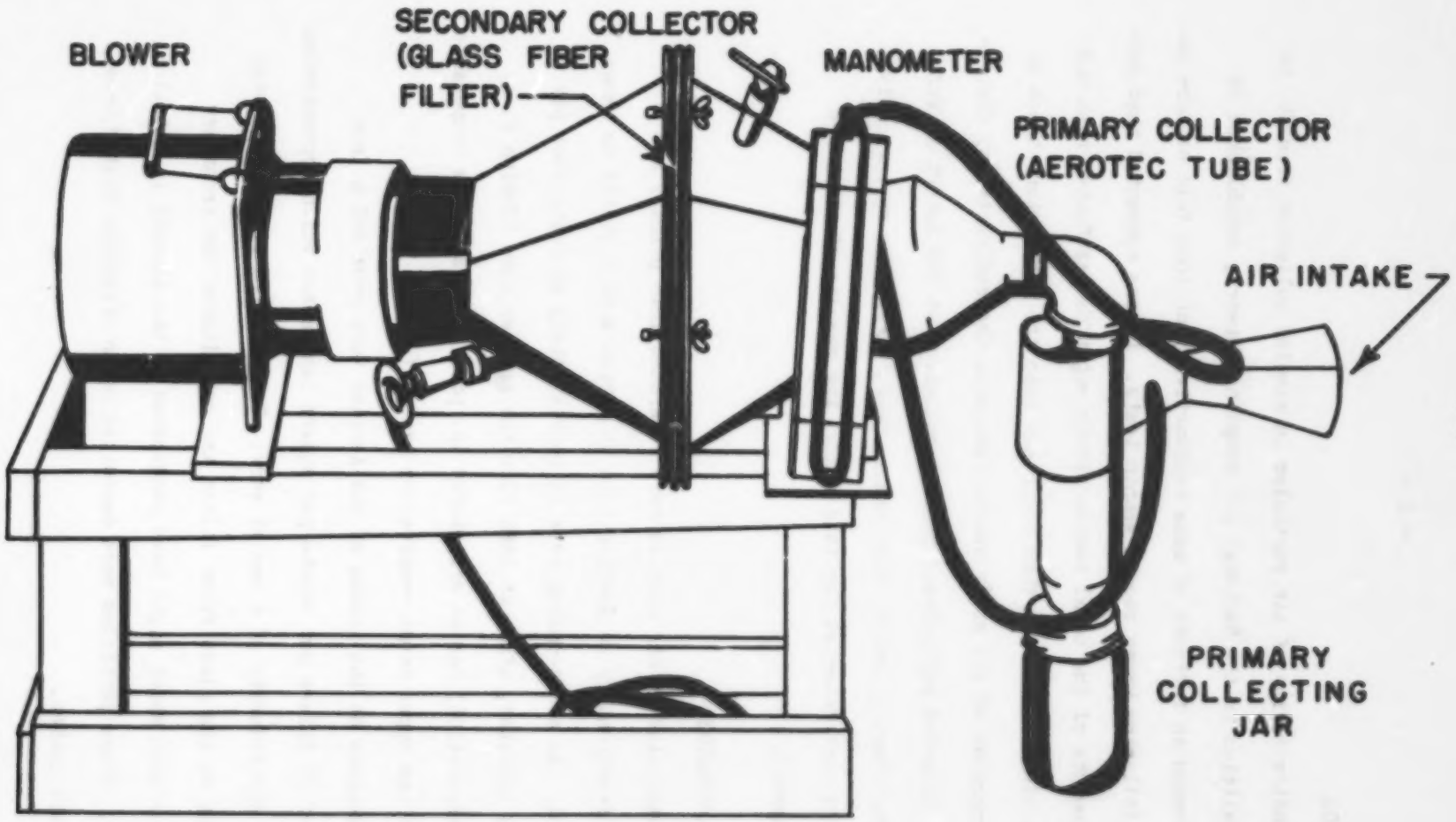


FIGURE I – AIR SAMPLER

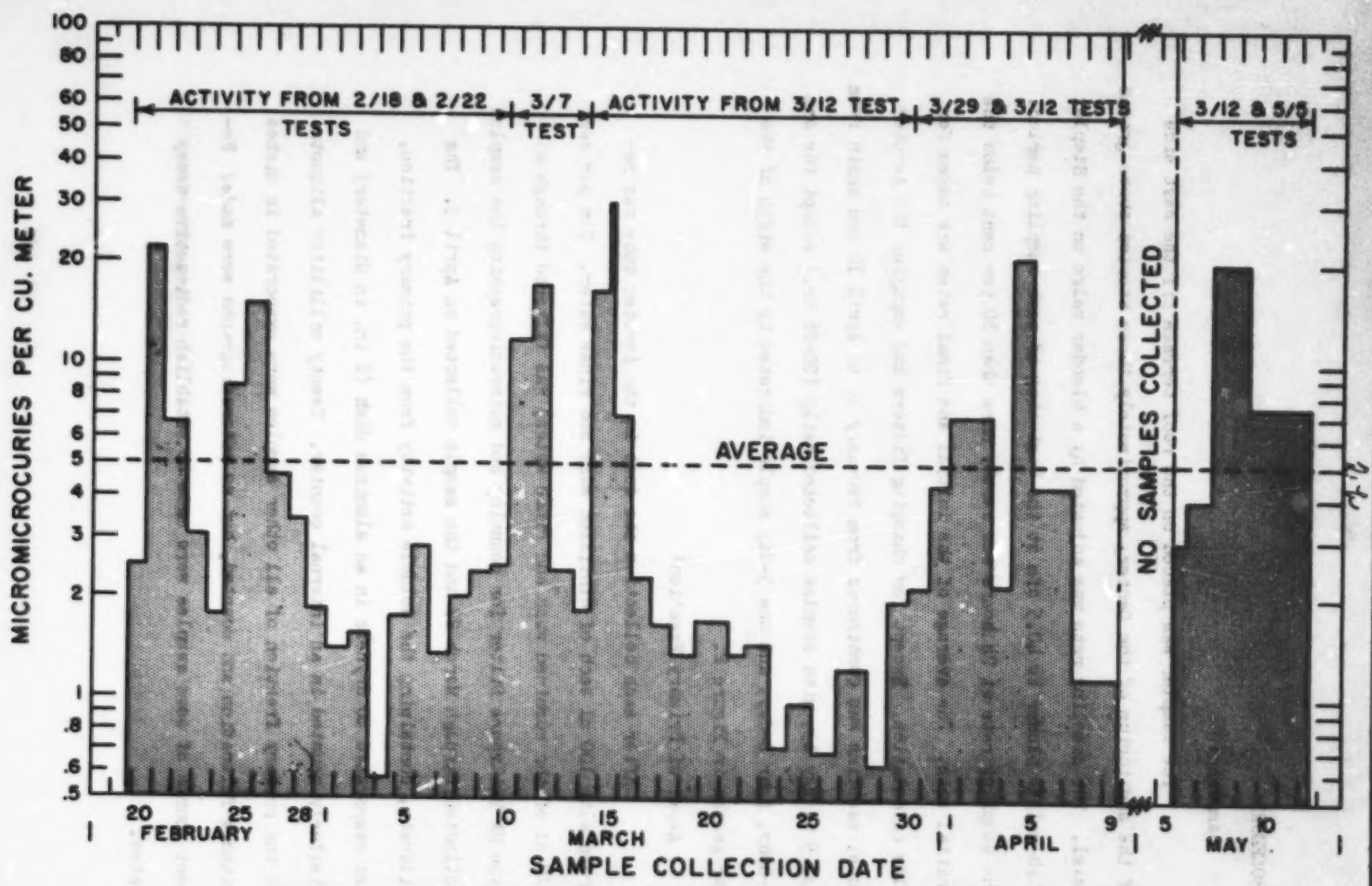
PROCEDURE

Sampling:

The sampler was placed on the roof terrace off the east side of the auditorium of the Center, approximately three stories above ground level. The sampling rate was adjusted by a bleeder valve on the Staplex high-volume blower to 40.5 cfm at the beginning of each sampling period. The sampling rate at 24 hours was rarely more than 10 per cent below the initial rate. The average of the initial and final rates was taken for flow calculation. Except for changing filters and emptying the Aerotec tube, sampling was continuous from February 19 to April 10 and again from May 5 to May 13, with samples collected daily (20-28 hr.) except for one 3-hour, four 2-day, and one 3-day sample indicated by the width of the bar graphs in Figure 2.

Assay of Primary Fraction[1]

After each collection the dust in the Aerotec tube was removed with 100 ml each of distilled wash and rinse water. Ten per cent (20 ml of the combined wash and rinse water) was filtered through a type HA Millipore filter for counting and autoradiographing the samples collected through March 22 and the sample collected on April 3. The filtrate, containing the soluble activity from the primary fraction, was evaporated to dryness in an aluminum dish (2 in. in diameter) and similarly counted in an internal counter. Twenty milliliter aliquots of the primary fraction of all other samples were evaporated in dishes without filtration and counted, but no autoradiographs were made. Repeat counts of some samples were made to establish radioactive decay rates.



BETA ACTIVITY OF ATMOSPHERIC PARTICULATES

FIGURE 2

Assay of Secondary Fraction:

A one and seven-eighths-inch diameter circle (1/23 of the total filter area) was punched from the center of the glass fiber filter, counted in an internal proportional counter, and used for autoradiography.

Beta Radioactivity:

The samples were held at least 4 and usually 16 or more hours after sample collection, to permit decay of radon daughter activity[4, 7] ($T_{1/2}$ = 38 minutes) before the beta-gamma mixed fission product activity was counted in an internal gas-flow proportional counter having an efficiency of 50 per cent[6]. The measured activity of a sample was corrected for decay to the mid-collection time from decay data or by the application of the exponential decay factor of -1.2 for fission-product activity formed on the most probable test date.

Autoradiography:

Six membrane or glass-fiber filter samples, 1 to 4 days after collection, were glued face up on a 5 x 7 inch sheet of paper. The sheet was then inverted on top of the emulsion of an eastman Kodak Blue Brand X-ray film. A sponge rubber pad mounted on a 5 x 7 inch piece of plywood was placed on the inverted paper and weighted with a 16 lb. brick. The film was usually exposed for 20 to 28 hours before development for normal contrast with standard Kodak X-ray chemicals according to the manufacturer's directions.

Calibration of X-ray Film:

A calibration of the optical density developed on X-ray film from exposure to fission-product particles was made by using an

isolated radioactive particle in the sample collected April 5, 1955. This particle of measured beta activity was used in a series of eight exposures ranging from 5 minutes to 19 hours and in distance of particle to emulsion from 40 to 500 microns. The density (transmission) profile of a particle autoradiograph was determined with the aid of a microphotometer which viewed a film area 46 microns in diameter by means of magnification (1:10) and an aperture mask on the photocell.

Photometer readings were taken progressively from the center of the irradiated area outward until the density faded into background fog. From these readings the radiation exposures at the points of measurement were calculated[5] and used to equate the total radiation exposure received by the film. Since the radioactivity and length of exposure were known, a proportionality constant could then be calculated for each of the eight calibration exposures. The average of the eight proportionality constants was used to calculate the radioactivity of air particulates from their autoradiographs. The calculated activity of a radioactive particle was then extrapolated from decay data to the activity at mid-collection time.

Autoradiographs of the more active particles, which had an activity at mid-collection time greater than about 5 micromicrocuries (μμc) were measured in this manner.

The minimum detectable activity by autoradiographic technique varies roughly as the square of the distance between the particle and the film emulsion. For a particle exposed for 24 hours and spaced one hundred microns from the emulsion the minimum detectable activity was estimated to be roughly one μμc. All particles having an

estimated radioactivity of less than 5 μμc were counted under a low power microscope and each was assumed to have an average activity of 2.3 μμc.

RESULTS

Gross Beta Activity *

The total beta radioactivity of particulate matter in each air sample is shown in Figure 2. The lower level of beta activity found in five daily samples during the study and two collections made a few days before the first U. S. nuclear weapons test in February 1955 ranged from 0.3 to 1 μμc per cubic meter (1 $\mu\mu c/M^3 = 10^{-12} \mu c/ml$). The remaining 43 samples had an activity greater than 1 but less than 30 $\mu\mu c/M^3$. Eight samples had an activity exceeding 10 $\mu\mu c/M^3$. The average activity of all samples was 5 $\mu\mu c/M^3$.

The highest levels of activity observed in Cincinnati appeared two to four days following the reported detonation of nuclear devices. Decay data of the air particulate samples and some rain samples were used to identify the radioactivity as having been formed from specific Nevada tests as shown in Figure 2.

The beta activity of particles in the primary (Aerotec) fraction ranged from 0.06 to 17, with an average of 1.7 μμc per cubic meter. The minimum value was found in a sample collected February 17. The activity of this sample had a half-life of several months. The maximum activity of a single particle was 1700 μμc, found in the April 5 sample. The particle was isolated and used for calibrating the autoradiographic technique.

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*Gross beta activity is counted at an efficiency of 50 per cent and includes gamma activity which is counted at an efficiency of 1 to 3 per cent.

Of the average activity of 5 μμc/M³ in air particulates, an average of 1.7 μμc/M³ or 34 per cent of the total was found in the primary fraction. Based on 28 samples available for comparison, from 1 to 41 per cent, or an average of 16 per cent, of the activity in the primary fraction was dissolved in the water used to remove the particles from the Aerotec unit.

Autoradiographic Assay:

Both the primary and secondary fractions of most of the samples collected between February 20 and March 19 and the sample collected on April 3 were autoradiographed after separating the water-soluble activity in the primary fraction. A typical autoradiograph of air particulates is shown in Figure 3. The autoradiographs revealed 106 active particles which were measured by photometry and 2,084 particles detected by microscopic examination.

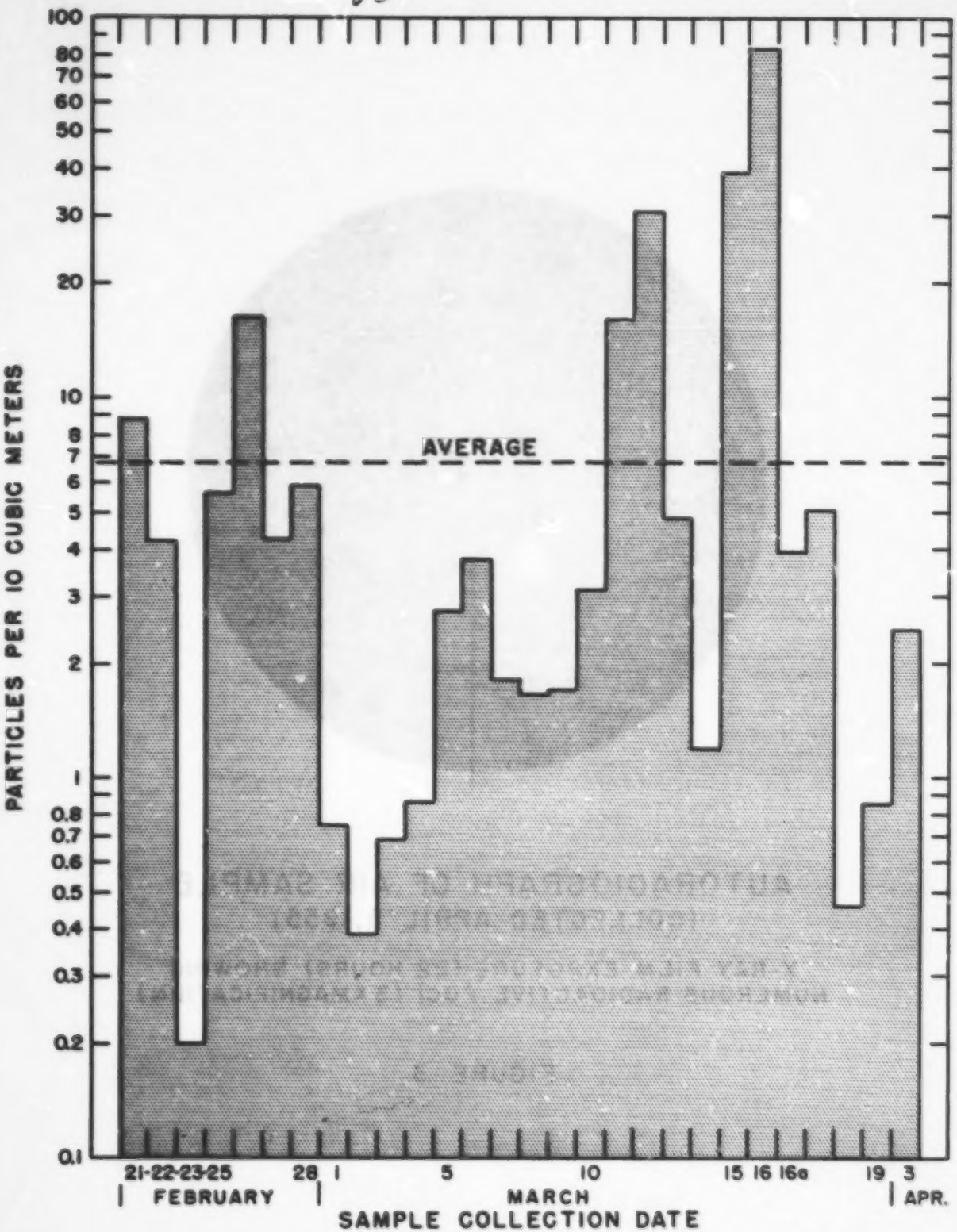
The number of radioactive particles of variable beta activity represented an extremely small percentage of the total particles in the air, but, when it is considered that a single person inhales 15 to 20 cubic meters of air daily, the particle radioactivity may have health significance. The variation in the number of radioactive particles in the 28 samples assayed by the autoradiographic technique is presented in Figure 4. Excluding the samples of February 23, 25, and March 1 for which only the primary fraction particles are given, the bar graph shows that all but 5 of 26 samples contained more than 1 radioactive particle per 10 cubic meters. In five samples there were from 16 to 82, as compared to a 26-sample average of 6.7 and a median of 4 radioactive particles per 10 cubic meters.

AUTORADIOGRAPH OF AIR SAMPLE
(COLLECTED APRIL 5,1955)

X-RAY FILM EXPOSURE (22 HOURS) SHOWING
NUMEROUS RADIOACTIVE FOCI (3 x MAGNIFICATION)

FIGURE 3

6c



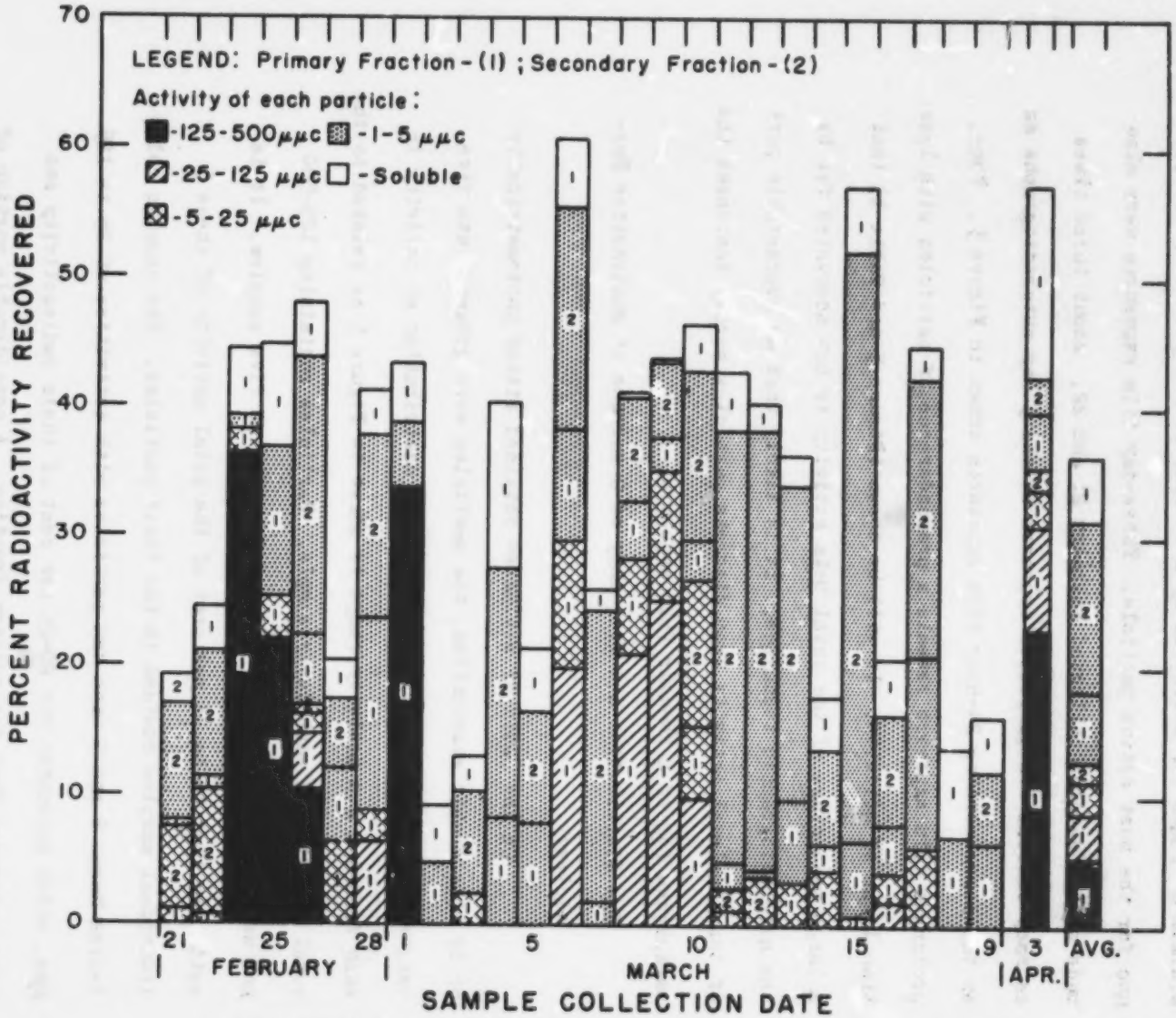
RADIOACTIVE PARTICLES IN CINCINNATI ATMOSPHERE

FIGURE 4

The beta activity of these particles ranged from an estimated minimum of 1 μμc for detection by a 24-hour film exposure up to 1,700 μμc for the most active particle. Three-day film exposures were also made on the samples collected February 21 and 22. About three times as many radioactive particles appeared on the 3-day autoradiographs as on the corresponding 24-hour film exposures shown in Figure 3. Thus, prolonged film exposure reveals a preponderance of particles with less than 1 μμc of activity. It will be shown (Figure 5 and Table 1) that a large percentage of the total beta activity is not accounted for by the autoradiographic technique. It is likely that an appreciable part of the low-level activity from discrete particles merely increases the background fog of a 24-hour film exposure.

The Distribution and Frequency of Occurence of Radioactive Particles:

On the basis of estimates obtained either photometrically or by microscopic examination, the particles were grouped into five ranges of beta activity. The percentage distribution of activity in each fraction and activity range is shown in Figure 5 as related to the total activity of each daily sample. Particles containing 125-500 μμc were found only in the primary fraction of five samples. It is estimated that 10 to 37 per cent of the total activity of these 5 individual samples resided in the "hot" particles. The samples collected March 8 and 9 each had particles with activities of 25 to 125 μμc, which accounted for 20-25 per cent of their radioactivity and were recovered from the primary fraction. A considerable portion of particles in the primary fraction also had activities of 1 to 25 μμc.



ACTIVITY OF PARTICLES BY AUTORADIOGRAPHS

FIGURE 5

The activity levels of particles in the secondary fraction were usually less than 5 μμc per particle, but 6 samples had secondary fraction particles having activities of 5 to 25 μμc. Figure 5 shows that at most only 61 per cent of the total activity of a sample was observed by the autoradiographic technique. This may be explained as follows: (1) the secondary fraction of three samples were not autoradiographed; (2) particles having an activity of less than about 1 μμc contributed only background fog to the X-ray films and could not be measured; (3) the assumed average activity of 2.3 μμc for particles having an activity of 1-5 μμc each may be an underestimate; and (4) the autoradiographic technique is at best a rough estimate of particle activity.

The results are further summarized in Table 1 to show the frequency of occurrence of active particles in the atmosphere tested. The most radioactive particle found (1700 μμc) had a diameter of only 7.5 microns. It appeared in the primary fraction and theoretically would occur at a frequency of 6 particles per 1000 cubic meters of air, whereas a maximum of 13 and an average of 1.5 particles having an activity of 125 to 500 μμc occurred per 1000 cubic meters of air. The great preponderance of active particles were found to have low activity levels of 1 - 5 μμc; they were much more abundant in the secondary fraction. There were no particles in the secondary fraction which had an activity exceeding 25 μμc.

Table 1 also shows that of the radioactivity in each fraction an average of 57 per cent was accounted for by autoradiography in the primary fractions and an average of 23 per cent was accounted for by autoradiography in the secondary fractions. Considering the activity

of both fractions, a maximum of 61, a minimum of 9, and an average of 36 per cent of the total activity was accountable by the autoradiographic technique.

Autoradiographs:

On autoradiographing 10 per cent of the primary fraction of the 24-hour sample collected at noon April 5, a particle having an estimated radioactivity of 1700 μμc was found. Subsequent autoradiographs of the entire primary sample failed to reveal any more particles of activity exceeding 500 μμc. The active particle was isolated from the mass of nonradioactive particles in the sample. It was a reddish, semi-transparent, nearly spherical particle having a diameter of 7.5 microns. Its decay and absorption data were characteristic of 8-day-old mixed fission product.

DISCUSSION

About one-third of the total radioactivity collected in Cincinnati air was of such particle size that if inhaled it would have been retained in the human upper respiratory system. The rest of the activity would have penetrated into the alveoli of the lungs. Presumably 50 per cent may have lodged in lung tissue for a variable time. Most of the particles in this fraction had an activity of less than 5 μμc per particle. However, some of the particles found in the primary fraction were much more radioactive and might have reached lung tissue. One active particle having a diameter of 7.5 microns was recovered from the primary fraction. Its size was such that, if inhaled, it would have had a finite chance of reaching the alveoli. Once there, the particle would

TABLE 1.

OCCURRENCE OF PARTICLES OF GIVEN RADIOACTIVITY

Particle activity range, μμc	Number of radioactive particles per 10 cubic meters of air			
	Primary fraction		Secondary fraction	
	Maximum	Average	Maximum	Average
1 - 5	8.35	1.17	73.5	5.2
5 - 25	0.38	0.12	1.5	0.16
25 - 125	0.13	0.025	0	0
125 - 500	0.13	0.015	0	0
1700	0.06*			
	Percentage of fraction activity in discrete particles**			
1 - 500	89	57	51	23

* One particle in 10⅝ of sample had a diameter of 7.5 microns and an activity of 1700 μμc.

** Based on autoradiographic technique employing 24-hour film exposure and, in the case of the primary fraction, its soluble activity which varied from 1 to 41 or an average of 16 per cent of the total primary activity.

probably remain to cause considerable radiation damage to local lung tissue. When inhaled the smaller particles are more likely to reach lung tissue but may be eliminated by several mechanisms. (1, 2, 8, 9)

SUMMARY

Radioactive particles found in Cincinnati air following nuclear detonations in Nevada were separated into two size-fractions representative of particles retained by the upper and the lower respiratory system. Air sampling was continuous from late February until early April and again for one week in May 1955.

Measurements by autoradiographs and gross internal proportional counting indicated that:

1. An average of 6.7 radioactive particles per 10 cubic meters of air was found in 27 samples collected between February 21 and March 19 and one sample collected on April 3. The weighted average beta activity of these samples was 37 μμc per 10 cubic meters of air. One sample contained as many as 80 radioactive particles and a beta activity of 300 μμc in 10 cubic meters of air.

2. The maximum, minimum, and average concentrations of beta activity in air from 47 samples collected between February 16 and April 9 and four samples collected between May 5 and May 12 were 30, 0.35, and 5 μμc per cubic meter of air, respectively.

3. The more radioactive particles were collected by a cyclone separator which simulates capture of particles by the nasal passages and bronchial tubes. Generally these particles would be rapidly

eliminated from the human system. In roughly half of the samples tested, these particles had a beta activity of 25 to 500 μμc, and one isolated particle of 7.5 micron diameter had an activity of 1700 μμc.

4. The activity of particles caught on the filter, representing material which would penetrate to the lung alveoli, was usually less than 5 μμc and did not exceed 25 μμc per particle.

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